Anisotropic thermal conductivity and permeability of solidified expanded natural graphite

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Abstract

Solidified expanded natural graphite (ENG) has good heat and mass transfer characteristics and has recently been utilized as a matrix for heat and mass transfer performance intensification in adsorption refrigeration and air conditioning equipment. In order to gain an overall understanding of the heat and mass transfer process in solidified expanded natural graphite, the anisotropic thermal conductivities and permeabilities are investigated in two types of solidified block, i.e. discs of solidified expanded natural graphite (DSENG) and plates of solidified expanded natural graphite (PSENG), in which the heat conductive and mass transfer directions are parallel and perpendicular, respectively, to the pressing direction producing blocks. An unexpected phenomenon is found in the research, which is that the thermal conductivity sometimes decreases while the density of the solidified ENG increases, this only occurring for the heat conductive direction perpendicular to the pressing direction. Results also show that the direction has larger thermal conductivity also has a better permeability; the phenomena of anisotropic thermal conductivities are strongly dependent on density. The reasons are analyzed in the paper, and they are mainly related with the distribution of micro layers inside the samples.

Keywords: Expanded natural graphite; thermal conductivity; permeability; micro layer

Introduction

Expanded graphite, which has good heat and mass transfer performance, recently has been kept utilized as a type of heat and mass transfer intensification matrix.

The most common such use of expanded graphite is for the development of new types of adsorbents for use in refrigeration or air conditioning. Mauran and Spinner first introduced expanded graphite in the adsorbent $CaCl_2$[1], and such a solidified composite adsorbent of $CaCl_2$ and expanded graphite was termed IMPEX. In this material a graphite bock was impregnated with a solution of 20% $CaCl_2$ [1,2]. Later, Han and Lee studied the permeability of expanded graphite-metallic salt composites for heat pumps, and found that the gas permeability was in the range of $5{:}0\times16^{-16}\sim10^{-12}$ m^2, depending on the reaction pair, bulk density and weight fraction of the graphite powder [3]. Fujioka developed a composite adsorbent composed of $CaCl_2$, expanded graphite, and activated carbon fiber, which improves the thermal conductivity of the adsorbent from less than 0.15 W/(mK) to over than 0.6 W/(mK) [4]. Recently Mauran's research group also proposed a type of energy storage system which involved $SrBr_2$ as reactant and H_2O as refrigerant fluid, with expanded graphite adopted as a heat and mass transfer intensification matrix [5]. Expanded graphite is also believed to be a promising material for heat transfer enhancement for adsorption hydrogen storage. For a pure ENG pellet with a porosity of 79.1%, the effective thermal conductivity of pure ENG pellets is about 8W/(mK) while the density of ENG is 1250 kg/m^3[6]. Compacted composites of $LaNi_5$ and recompressed expanded graphite have been made and improved the thermal conductivity of the hydride from 0.1W/(mK) to 3–6 W/(mK) showing a good potential for the development of high power metal hydride devices [7]. In gas separation, expanded graphite had been utilized as an additive for the granular adsorbents of activated carbon, in which the thermal conductivity of activated carbon is improved by over than 20 times [8]. Expanded graphite has also been utilized for the heat transfer intensification of phase change materials, such as paraffin, in which the thermal conductivity was improved from 0.22 W/(mK) to over than 0.8W/(mK) [9].

The main characteristic of composite adsorbents with a graphite matrix is the anisotropic heat transfer performance, which was first found by Han. Han et al had studied expanded graphite matrices for heat pump [10,11], and they reported the anisotropic heat transfer behavior of solidified adsorbent with expanded graphite matrix [11] in 1996, pointing out the direction perpendicular to the pressing direction for solidification of the adsorbents always has the higher thermal conductivity. This conclusion is now generally accepted by researchers. Recently non-uniform reactive blocks were manufactured by the impregnation of metal salts into the expanded graphite to improve the heat and mass transfer ability in chemical heat pumps. The experimental results showed that such a block can improve the heat transfer properties and additionally overcome the mass transfer limitation due to high apparent density of the block which diminishes the permeability of refrigerant (ammonia) gas [12]. But there are many fewer experimental results for anisotropic thermal conductivities and similarly few studies on the anisotropic permeabilities, which are very essential to the understanding of the whole heat and mass transfer performance.

The main problem in research on the solidified adsorbent with expanded graphite as matrix is the big difference for the values of thermal conductivities that are tested by different researchers. Mostly the value is lower than 6W/(mK), but for the solidified adsorbents that react with water and utilizes graphite as matrix, such as silica gel [13] and $CaCl_2$ [14] were tested in the ambient temperature, and the thermal conductivity could be as high as 9-10W/(mK). Recently, the same graphite block with the same density of the matrix in reference [14] without $CaCl_2$ was produced, and its thermal conductivity was only about 2~3W/(mK), much lower than the previous research results [14]. The reason for this anomaly is analyzed, and it is suggested that it is caused by the combination of the use of unsteady conductivity test methods when adsorption heat may be present. Hydrophilic adsorbents will react with the water in the air while its thermal conductivity is tested in the ambient environment. In such conditions the temperature difference between measuring point and heating point will be less than the condition without reaction heat, and then the thermal conductivity tested will be much larger than the normal values.

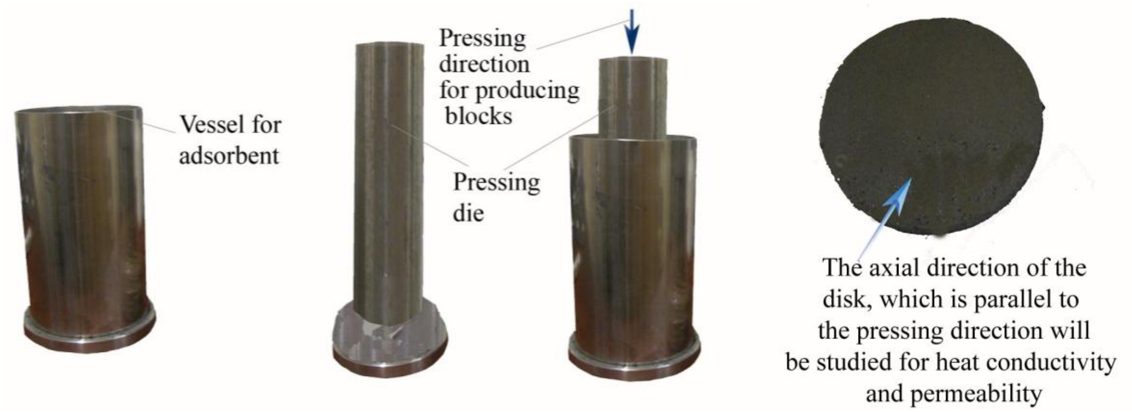
In order to give an provide an overview on the anisotropic thermal conductivity and permeability differences for the expanded graphite matrix and at the same time eliminate the errors that could be caused by hydrophilic adsorbents, the thermal conductivities and permeabilities of solid blocks of pure expanded natural graphite with different pressing methods are studied.

1 Development of the solidified blocks

The expanded natural graphite (ENG) is prepared by heating untreated natural graphite, which is 100 mesh and manufactured in China, in an oven at the temperature of 700^{o}C for 12~15 minutes. In order to obtain the solidified blocks for which the anisotropic thermal conductivities and permeabilities could be measured, two types of rigs were used.

1.1 The rig for the disc solidified ENG blocks (DSENG)

The rig for the disc solidified ENG blocks (DSENG) is shown in Fig.1a, which is mainly for the production of the disc blocks in which the thermal conductivity and permeability will be measured parallel to the pressing direction. The procedures to produce the blocks involves putting the adsorbent inside the vessel, and then pressing the adsorbent with a force of 0~100kN applied by the die in a press. The DSENG block produced is shown in Fig.1b. The thermal conductivities and permeabilities in the axial direction of the disc, which is parallel to the pressing direction, were studied.



(a) (b)

Fig.1 (a) The rig for the disc solidified ENG blocks, (b) disc solidified ENG block

1.1 The rig for the plate solidified ENG blocks (PSENG)

The rig for the plate solidified ENG blocks is shown in Fig.2, which is mainly for the production of the blocks in which conductivities and permeabilities perpendicular to the pressing direction could be measured.

The block of PSENG is prepared by the rig in Fig.2 is shown in Fig.3a. For the measurement of properties the PSENG was cut into a circular shape (Fig.3b) that is required by the thermal conductivity and permeability test equipment. Then the thermal conductivities and permeabilities in the direction that is perpendicular to the pressing direction could be tested.

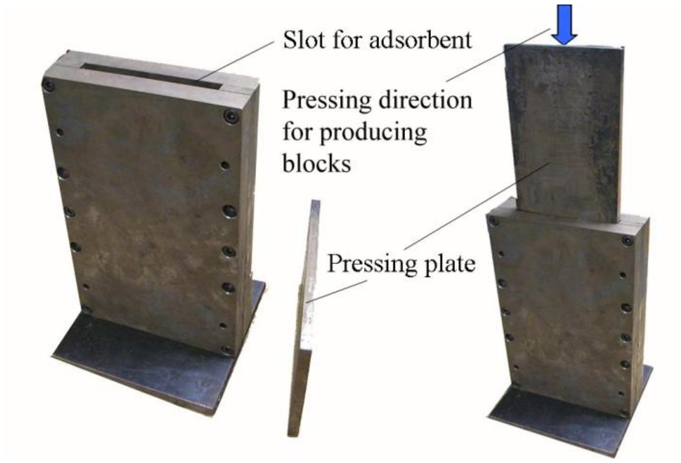
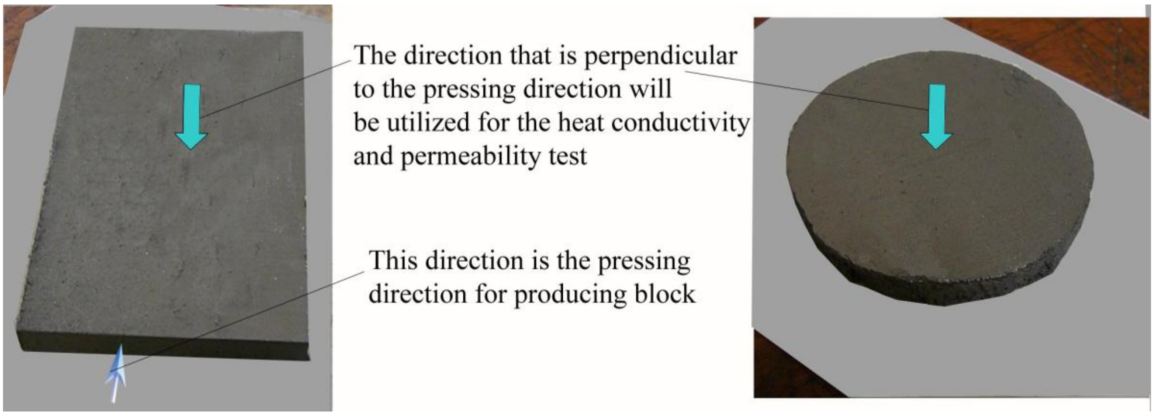


Fig.2 The rig for the plate solidified ENG blocks



(a) (b)

Fig.3 Block of PSENG (a) plate sample; (b) circle sample cut from plate sample for thermal conductivity and permeability research

2 Researches on anisotropic thermal conductivities

2.1 Thermal conductivity test unit

The thermal conductivity is investigated by using the guard-hot plate method [15]. It is an absolute method for determining the steady state thermal conductivity of materials. The experimental set-up is based on British Standard BS-874 [16]. The photo of the test unit is shown in Fig.4. The main components are: a central disc heater sandwiched between two solidified ENG blocks, two water coolers symmetrically above and below the central disc heater, an annular guard heater radially beyond the central disc heater, and a water tank in which the water is pumped to the circuit linked with coolers. Copper blocks serves as the clamping force on the whole test unit. The test unit is thermal isolated before experiments.

The determination of the effective thermal conductivity λ is based on the measurement of the average temperature gradient ΔT produced through the solidified blocks by a known axial heat flux Q under steady-state conditions. When the working conditions (heat flux determined by the electric current of central disc heater, water flow rate, temperatures) are set up and the equilibrium is reached, the effective thermal conductivity λ (W/(mK)) is given by the following expression:

$$\lambda = \frac{Q \times \Delta z}{2S\Delta T} \tag{1}$$

where Q is the measured central disc heater heating power (W), Δz is the thickness of the solidified expanded graphite blocks, ΔT is the temperature difference across the carbon sample and S is the effective heating area of the central plate heater (m^2).

The intrinsic thermal conductivity is determined by combining the contact solidified ENG block-aluminum heat transfer coefficient and the total effective thermal resistance of the sample deduced from the experiments, which are shown in reference [15].

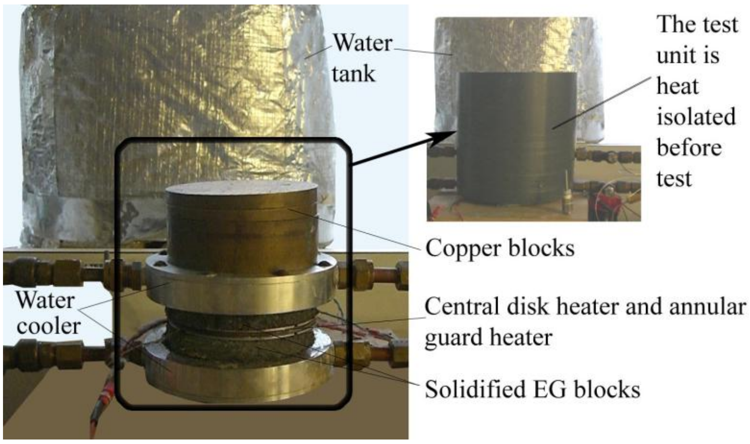


Fig.4 The thermal conductivity test unit

2.2 The thermal conductivity of DSENG

Disc solidified expanded natural graphite (DSENG) of different densities were produced, and the thermal conductivities measured by the test unit in Fig.4. The thermal conductivities are tested for a range of electric currents of the central heater. The values of the thermal conductivity measured varies little with the electric current of central heater as is shown in Fig.5a but varies considerably from sample to sample. The average thermal conductivities under the condition of different currents are calculated and the relation between thermal conductivities and densities are shown in Fig.5b. Fig.5b shows an interesting phenomenon, which is that there is a range where the thermal conductivity decreases while the density increases. In Fig.5b while the density of the solidified disc is lower than 300 kg/m^3, the thermal conductivity increases while the density of the disc increases. The values of thermal conductivity vary very slightly while the values of the density of the discs range from 343kg/m^3 to 576kg/m^3, and the optimal thermal conductivity, which is 1.70W/(mK), is also obtained in this range. The thermal conductivity decreases when the density of the adsorbent is higher than 658kg/m^3, and the lowest thermal conductivity of 1.21W/(mK) is obtained when the density of the adsorbent is 698kg/m^3. After that the thermal conductivity increases again while the density increases, and the value is 1.318W/(mK) for 730kg/m^3.

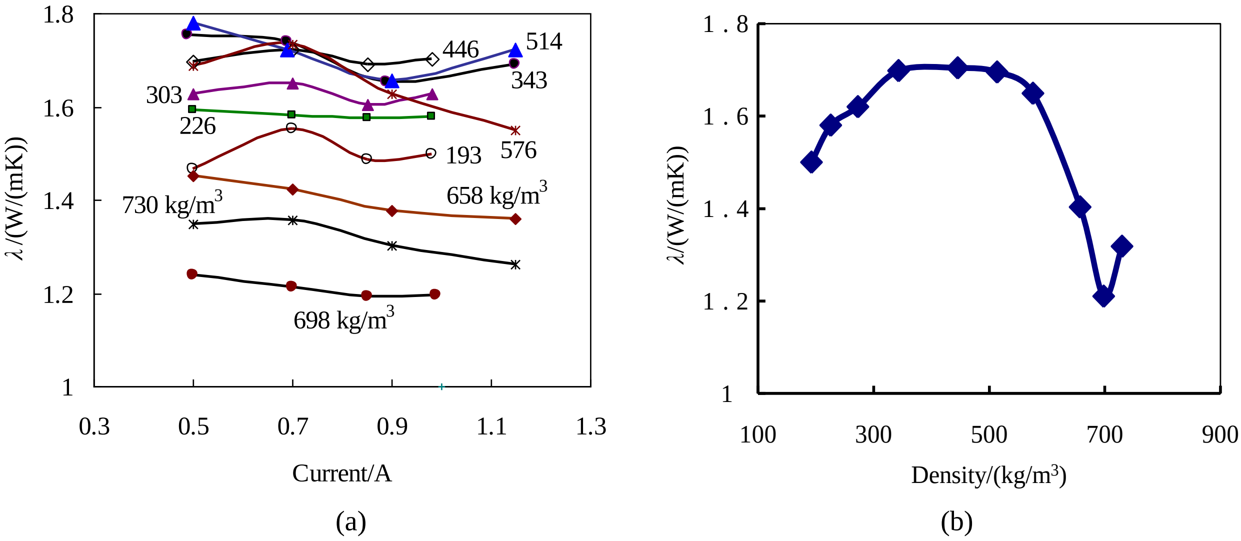


Fig.5 The thermal conductivities of DSENG, (a) thermal conductivities vs. electric current of central heater, (b) average thermal conductivity vs. densities

Generally for any type of material, the thermal conductivities always increase while the densities of the material increase. In order to find the reason for this abnormal phenomena of DSENG, scanning electron microscope (SEM) pictures are taken of different samples, as are shown in Fig.6.

For the expanded graphite, the micro layers which are perpendicular to the pressing direction will be formed as the solidified adsorbent is produced. For the DSENG the heat conductive direction is parallel with the pressing direction, i.e. the heat conductive direction will be perpendicular to most micro layers. Fig.6a and 6b shows that when the density is lower, for example, when the density of the solidified DSENG is 343kg/m^3, the micro layers inside the sample distribute disorderly because the pressing force to make the DSENG with less density is less. Such a structure is helpful for the heat conductive process because the layers that are parallel to the heat conductive directions exist in the sample. When the density of the sample increase, for example, when the density is 446kg/m^3 (Fig.6c), although the layer began to distribute more uniformly along the horizontal direction and some perpendicular layers are destroyed by the larger pressing force, there are also some perpendicular layers remaining, thus the thermal conductivity in Fig.5 is also useful. But when the density of the adsorbent is larger than 658kg/m^3, from Fig.6d, 6e and 6f we can see that the layers are distribute uniformly along the horizontal direction, and the perpendicular layers that are essential for the heat conductive process in the perpendicular direction are disrupted seriously by the larger pressing force, thus the thermal conductivity in the perpendicular direction decreases although the density increases. After most of the perpendicular layers are destroyed, the thermal conductivity will increase again while the density increases, that is mainly for the reason of that the heat resistance between horizontal layers decreases for the reason of the larger density that produced by larger pressing force.

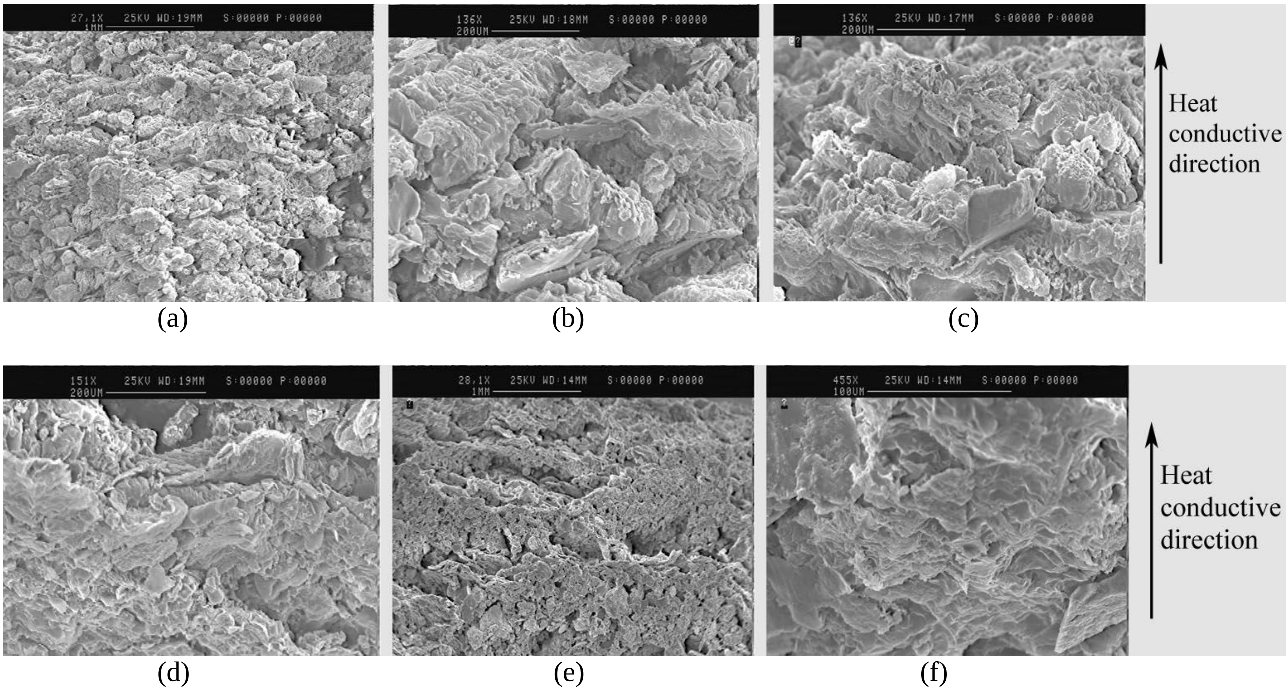


Fig.6 SEM pictures of DSENG with different densities. (a) 343kg/m^3, 27.1X, (b) 343 kg/m^3, 136X, (c) 446kg/m^3, 136X, (d) 658kg/m^3, 151X, (e) 730kg/m^3, 28.1X, (f) 730343 kg/m^3, 455X

Fig.6d shows that the micro layer distributed very uniformly. For such a structure the thermal conductivity will not be good in the direction perpendicular to the layers, but the optimal heat transfer performance will be obtained on the direction parallel to layers. In order to research such conditions, the plate solidified expanded natural graphite (PSENG) is studied.

2.3 The thermal conductivity of PSENG

The samples of plate solidified expanded natural graphite (PSENG) with different densities were produced. The samples were cut into circular shapes, and then the thermal conductivities were measured. The experimental results are shown in Fig.7. Fig.7a is the relation between thermal conductivity and the current, and it shows that the thermal conductivity varies very slightly while the electric current of the central heater in Fig.4 changes. Fig.7b is the relation between average thermal conductivities for each sample. It can be seen that the thermal conductivity keeps increasing with increasing density.

The trend of the thermal conductivities in Fig.7b is analyzed, and it is also related to the distribution of the micro layers inside the samples. The SEM pictures of PSENG are shown in Fig.8. For PSENG the heat conductive direction tested by the test unit is perpendicular to the pressing direction. For such a condition, because the layers of the expanded graphite are also perpendicular to the pressing direction, the heat conductive direction is parallel with the distribution of layers, just as the Fig.8a and 8b shows. The layers are distributed more uniformly while the density that is pressed by larger force is larger, thus the thermal conductivity is higher (Fig.7b) while the density of the sample increases.

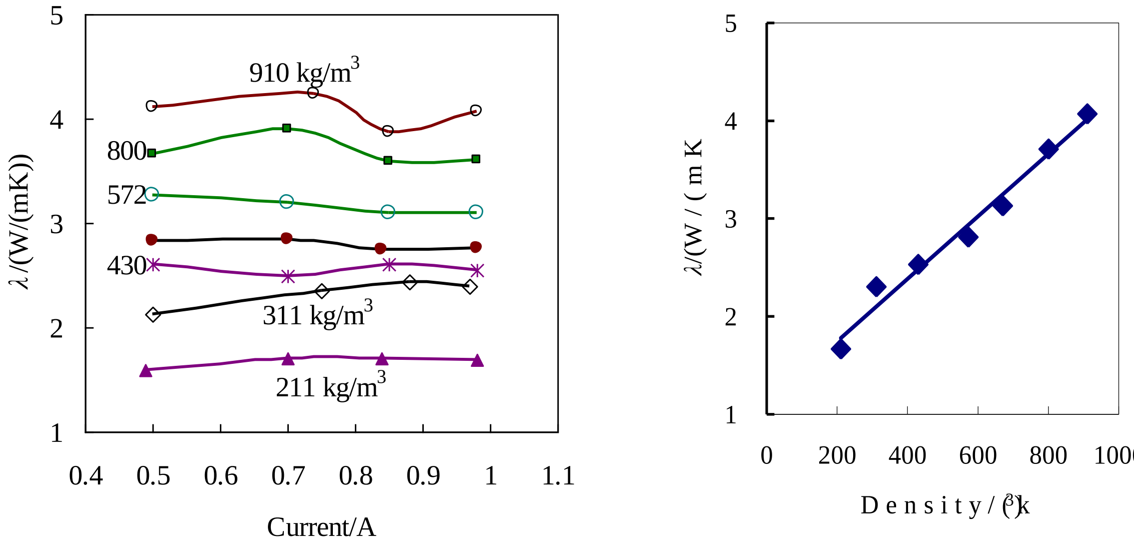


Fig.7 The thermal conductivities of PSENG, (a) thermal conductivities vs. electric current of central heater, (b) average thermal conductivity vs. densities

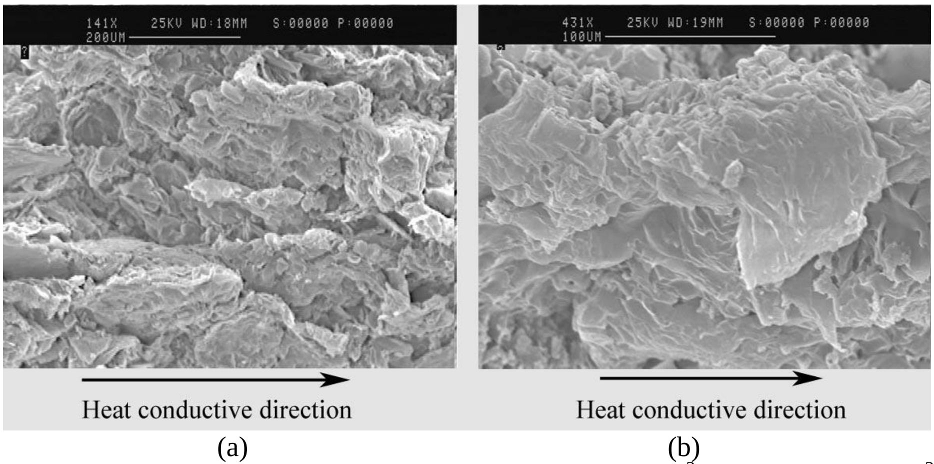


Fig.8 SEM pictures of PSENG with different densities, (a) 557kg/m^3, 141X, (b) 700kg/m^3, 431X

The results in Fig.5b and Fig.7b are compared, the PSENG has a much larger value of thermal conductivity than that of the DSENG when the densities of these two types of blocks are similar, and the difference is larger while the density is higher. For example, when the density is between 210~220kg/m^3, the thermal conductivities of PSENG and DSENG are 1.67 and 1.58W/(mK), respectively, the value of PSENG is only improved by 6%. When the density is about 660~670 kg/m^3, the thermal conductivities of PSENG and DSENG are 3.13 and 1.40 W/(mK), respectively, the value of PSENG improved by about 2 times. Such phenomena are also related with the micro layers distribution, which are disorderly for smaller density and uniform for larger density, and there will be higher heat transfer resistance for the direction perpendicular to the distribution of layers at large density.

3 Anisotropic permeability

The micro layers inside the solidified expanded graphite not only influence the thermal conductivity, but will also influence the permeability.

The permeability of DSENG and PSENG were tested in order to study the anisotropic variation of permeability in the solidified expanded graphite.

3.1 The permeability test unit

The permeabilities of the samples are tested by using a specially designed test unit shown in Fig.9 [15]. The main components are the sample holder, pressure drop meter, and a rotameter. The permeability test comprises measurements of the pressure drop Δp across the sample of block disc when compressed air is flowing through it with flow rate q_v. Since the samples to be tested are porous media with very low gas velocities, the Ergun model is applicable. Assuming the gas used is ideal and that there is no mass accumulation inside the sample, for the axial gas flow configuration, the intrinsic characteristics of the material is then given by the following expression [15]:

$$W = \frac{1}{K} + BX \qquad (2)$$

In which

$$W = \frac{(p_1^2 - p_2^2)S}{2RT\mu m_a \Delta z}; \qquad X = \frac{m_a}{\mu S}; \qquad m_a = \rho S v_a \qquad (3)$$

where K is permeability (m^2), B is the shape factors of the samples, p_1 and p_2 are inlet pressure and outlet pressure of pressure air, and they can be gotten by the outlet pressure and the pressure drop measured. S is sample cross section (m^2), R is the gas constant (J/(kg K)), T is the sample temperature (K), which doesn't change significantly through the sample, μ and ρ are the gas viscosity (Pa s) and density (kg m^{-3}), respectively; m_a is gas mass flowrate (kg/s), v_a are axial velocities (m/s).

To measure the permeabilities, W and X are calculated by using the experimental data of flowrate, pressure drop, ambient temperature, and outlet gas pressure, etc., and then K is obtained from the relations between W and X, which is linear and $1/K$ is the intercept of the equation for the linear relation between them.

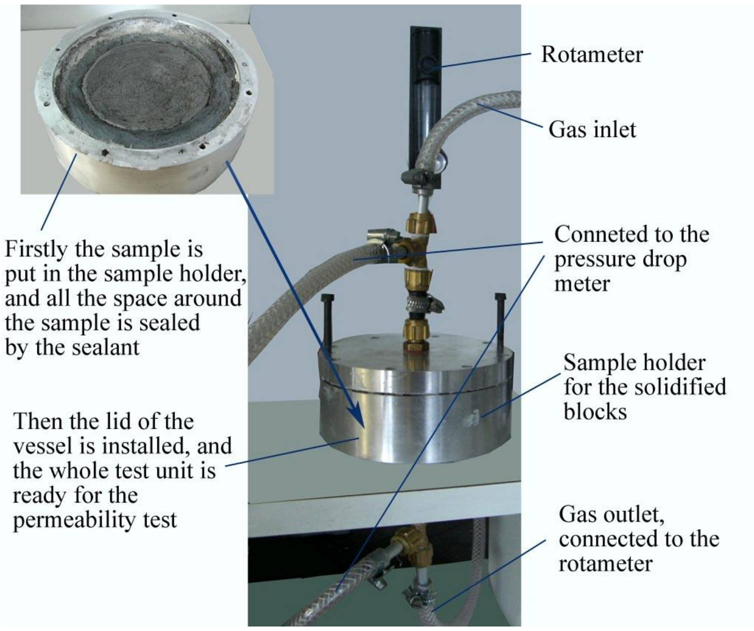


Fig.9 The permeability test unit

3.2 Experimental results

Four types of blocks are chosen for the comparison of the anisotropic permeability researches, which are two blocks of PSENG and DSENG, respectively, with the similar density of 430~450kg/m^3, and another two blocks with the similar density of 650~700kg/m^3.

The experimental results of the relations between gas flow rate (q_v) and pressure drop (Δp) are shown in Fig.10. The values of permeability are calculated by the experimental results in Fig.10, and they are shown in Table 1. In Table 1 we can see that for the same type of blocks, the permeability decreases while the density of the block increases. For the different type of blocks with similar densities, the PSENG has higher permeability than that of DSENG. For example, when the samples with similar density of about 440kg/m^3, the permeability of

PSENG is higher than 3 times of that of DSENG. It is mainly related with the micro layers inside the sample, which are shown in Fig.6 and Fig.8. For DSENG because the mass transfer direction of the gas is perpendicular to the distribution of most micro layers, the uniform layers will have a larger resistance for the mass transfer process, and then the permeability will be lower. For PSENG the mass transfer direction of the gas is parallel to the distribution of the micro layers, thus the situation is much different.

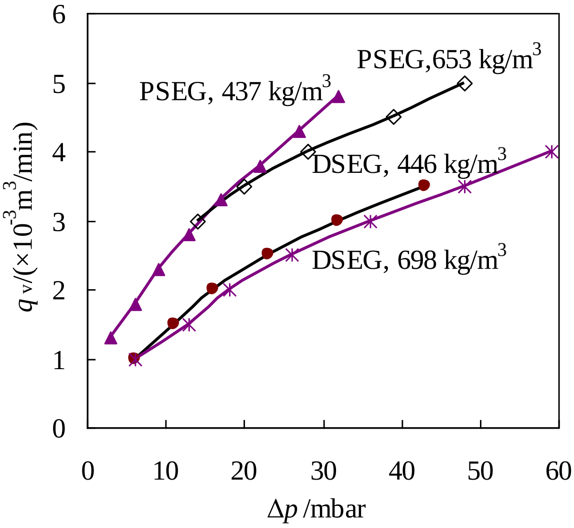


Fig.10 The values of gas flow rate vs. values of pressure drop

Table 1 The values of permeability of different samples

The type of blocks	Density (kg/m^3)	Permeability (m^2)	Characteristics of gas transfer direction
DSENG	446	2.076×10^{-12}	Parallel to the pressing direction and perpendicular to the micro layers
	698	1.873×10^{-12}	
PSENG	437	8.788×10^{-12}	Perpendicular to the pressing direction and parallel to the micro layers
	653	4.495×10^{-12}	

4 Conclusions

Disc solidified expanded natural graphite (DSENG) and plate solidified expanded natural graphite (PSENG), which has the heat conductive and gas transfer direction parallel and perpendicular, respectively, to the pressing direction producing blocks, were produced, and the anisotropic thermal conductivities and permeabilities of solidified expanded graphite were studied. Several new phenomena and conclusions are obtained as follows:

(1) For expanded graphite, its thermal conductivity sometimes will decrease while the density increases. Such a phenomenon occurs only in the heat transfer direction that is parallel to the pressing direction producing blocks, i.e. DSENG. This phenomenon is analyzed by using SEM pictures, and it is mainly caused by the arrangement of micro layers in the samples. The micro layers are distributed disorderly inside the sample when the density of the sample is smaller, and the layers that are parallel to the heat transfer direction help the heat conductive process. For the samples with larger density, the micro layers that are perpendicular to the pressing direction will distributed uniformly, and the micro layers that are parallel to the pressing direction, i.e. parallel to the heat transfer direction and helpful for the heat transfer process, are destroyed by larger pressing force, thus the thermal conductivity decreases although density increases. After most micro layers parallel to the heat conductive direction are destroyed, the thermal conductivity will increase again while the density increases; this is mainly because of the reduced heat resistance between the micro layers perpendicular to the heat conductive direction at larger densities.

(2) For a block of PSENG, because its heat conductive direction is perpendicular to the pressing direction used to produce the blocks, i.e., its heat conductive direction is parallel to the micro layers distribution, its thermal conductivity increases while the density increases. Its thermal conductivity is higher than 4W/(mK) when the density is about 910kg/m^3.

(3) The anisotropic heat conductive performances also related to the density, and it is more pronounced for the larger density of solidified blocks. When the density of the blocks are as low as 210~220kg/m^3, there is much less difference between the thermal conductivities of PSENG and DSENG, but when the density is higher than

660~670kg/m^3, the thermal conductivity of the PSENG is about 2 times of that of DSENG. These phenomena are also related to the micro layer distribution, which is disordered for smaller density and uniform for larger density, and it has higher heat transfer resistance in the direction perpendicular to the distribution of layers for large density.

(4) For most materials, the direction has higher thermal conductivity always has less gas permeability, but for the blocks of solidified expanded graphite, the situation is the inverse. The direction that is perpendicular to the pressing direction producing blocks not only has higher thermal conductivity, but also has better gas permeability, which is similar with the monolithic carbon [15]. For example, when the densities of the blocks are about 440kg/m^3, the thermal conductivities of PSENG and DSENG are 2.53 and 1.70 W/(mK), the value of PSENG is 49% higher than that of DSENG, meanwhile the gas permeability of PSENG and DSENG are 8.788×10^{-12} m^2 and 2.076×10^{-12} m^2, respectively, the value of PSENG is 3 times higher than that of DSENG. Such a phenomenon is also related with the distribution of micro layers. PSENG has a heat conductive and gas transfer direction parallel to the micro layer distributions, such a structure not only helpful for the heat transfer process, but also has a less resistance for the mass transfer process.

(5) For a same type of blocks, such as for DSENG or PSENG, the permeability decreases while the density increases. It is mainly caused by the reduced volume of mass transfer pores inside the sample with larger density.

The anisotropic heat conductive and mass transfer phenomena are essential for the development of new type materials with expanded graphite as matrix. The heat and mass transfer directions should be kept perpendicular to the pressing direction for the production of new materials. For some situations if the heat and mass transfer direction parallel to the pressing direction is the only choice, then for the development of the new type materials the density with optimal thermal conductivity should be studied.

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